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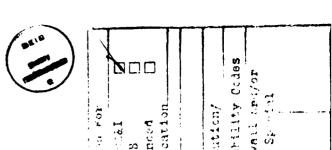
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in laminated UHC steel composites manufactured through solid state bonding. The unique mechanical properties achieved in UHC steels are due to the presence of micron-size ferrite grains and ultrafine spheroidized carbides.

ULTRAHIGH CARBON STEELS

by

O. D. Sherby, T. Oyama, D. W. Kum, B. Walser and J. Wadsworth[*]

SUMMARY

Recent studies and results on ultrahigh carbon (UHC) steels would suggest that major development efforts on these steels are timely and that programs to evaluate prototype structural components should be initiated. These recent results include: the development of economical processing techniques incorporating a divorced eutectoid transformation, the improvement of room temperature strength and ductility by heat treatment, the enhancement of superplastic properties through dilute alloying with silicon, and the attainment of exceptional notch impact strength in laminated UHC steel composites manufactured through solid state bonding. The unique mechanical properties achieved in UHC steels are due to the presence of micron-size ferrite grains and ultrafine spheroidized carbides.

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ULTRAHIGH CARBON STEELS

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Introduction

In the mid 1970's it was demonstrated that a class of steels now known as ultrahigh carbon (UHC) steels could be made to behave superplastically.[1-5] The steels are plain carbon steels containing 1.0 to 2.1% carbon (fifteen to thirty-two volume percent cementite). Traditionally, steels of this high carbon content have been neglected by industry because of a belief that they are inherently brittle (although these steels do have a rich and fascinating history because of their similarity in composition to Damascus steels[6-9]). UHC steels are now considered to have technological potential because when processed to develop ultrafine ferrite grains, 0.5-2 µm, containing fine spheroidized cementite particles, they have been shown not only to be superplastic at intermediate temperatures, but also to be strong and ductile at room temperature.[10-16] Further, because of their high carbon content, these steels can be made very hard (R_C = 65-68) by appropriate heat treatment after processing.[17] Fine grained UHC steels can also be solid state diffusion bonded readily either to themselves, or to other ferrous base materials, at temperatures below the A_1 temperature.[18] This unique ability has been utilized to prepare ferrous laminated composites with superplastic properties at intermediate temperatures[19-21] and with very high impact resistance at low temperatures.[21-23] In addition, the concepts de-

veloped in solid state bonding of UHC Steels have been utilized to prepare compacted powders of white cast iron (2 to 3%C) which exhibit superplastic behavior at 650 ^{O}C and which are ductile in tension at room temperature.[24-29]

In the past three years a number of significant new advances in the processing and properties of UHC steels have been made and it is the purpose of the present paper to review their current status.

1. Superplastic Formability of UHC Steels

The original work on UHC steels demonstrated that superplasticity could be developed over the range of composition from about 0.8 to 2.1%C and the range of temperature from 650 to 800°C.[12,13] This temperature range is from just below the A_1 temperature (727°C) to just above it. Furthermore, superplasticity was observed at intermediate strain rates (10^{-5} to 10^{-3} s^{-1}). Two new directions have been recently explored which have expanded the usefulness of superplasticity in UHC steels. The first relates to a novel alloying concept which extends the temperature and strain rate range for superplastic flow of UHC steels.[30,31] The second relates to novel processing methods[25-29] which have increased the range of carbon composition over which superplasticity can be observed. An overview of the expansion in composition and temperature ranges now available for superplasticity in UHC steels and cast irons is shown in the iron-cementite phase diagram in Figure 1.

1.1 Alloying to Improve Superplasticity in UHC Steels

The potential for commercial application of superplastic forming of UHC steel would be enhanced if superplasticity could be achieved at strain rates

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higher than $10^{-3}s^{-1}$. There are two methods of achieving such a goal: (1) by maintaining a fine grain size but increasing the temperature of metal forming, or (2) by decreasing the grain size, thereby making superplastic flow by grain boundary sliding at a given temperature more facile. It was discovered that the first method could be utilized in UHC steels by dilute alloying with silicon.[30,31]

Silicon was chosen as an alloying addition that could alter the superplastic properties of UHC steels because of its influence on the thermodynamics of the Fe-C system and on the kinetics of carbide formation and dissolution. Specifically, it is known that Si is a ferrite stabilizer and therefore increases the A_1 temperature of Fe-C alloys; this increase results in a wider ferrite temperature range over which superplastic flow is possible by comparison with plain carbon UHC steels. Furthermore, because the carbon content of the eutectoid composition is decreased with Si additions, Si increases the quantity of proeutectoid carbides. For this reason, there is an increase in the volume fraction of carbides available for grain boundary pinning in the austenite plus cementite range. Superplastic flow in austenite can thus now be expected, a property difficult to achieve in plain carbon UHC steels because rapid grain growth occurs in austenite. Thus the temperature range for superplastic flow is predicted to be increased with UHC-Si steels.

In addition to improving the temperature range over which superplastic flow should be obtained, Si also inhibits grain growth of ferrite grains during superplastic deformation below the A_1 temperature. This is because Si inhibits carbide particle growth and hence ferrite grain growth (which is controlled principally by carbide particle coarsening) is reduced. Another benefit of Si is that it dissolves in the ferrite or austenite matrix of iron. The hardness of the carbides is therefore unaffected since Si does not con-

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tribute to the structure of the carbides. An increase in hardness of the carbides could be deleterious to superplasticity.[32] As a solid solution strengthener, however, Si can be expected to make slip processes more difficult during superplastic deformation, thereby extending the range of strain rate over which grain boundary sliding (superplastic flow) dominates deformation.[32]

To illustrate the above principles, a UHC steel containing 1.25%C, 3%Si, 1.5%Cr and 0.5%Mn (henceforth designated as UHC-3Si steel) was thermomechanically processed to contain a fine grain size. The processing details are given in the next section. The final microstructure revealed a ferrite grain size of about 1 m with spheroidized carbide particles of 0.1 to 1 μm in diameter.

Figure 2 illustrates the log strain rate-log stress relation for the UHC-3Si steel at 800°C. At this temperature, the UHC-3Si steel is still in the ferrite range. The slope of the curve is the stress exponent, n, i.e., the reciprocal of the strain rate sensitivity exponent, m. As can be seen, the strain rate sensitivity exponent is high over the range of strain rates tested (m = 0.5). Even at a strain rate of 10^{-2} s^{-1}, m is about 0.4. A UHC steel with only 0.5% Si (1.5%C - 1.5%Cr - 0.5%Mn - 0.5%Si - balance Fe) is shown on Figure 2 for comparison. This UHC steel is not superplastic at 800°C, exhibiting a relatively low strain rate sensitivity exponent of 0.3. The reason for this latter behavior is that the low-Si UHC steel, at 800°C, is in the austenite plus cementite range and, because of the presence of only a small amount of undissolved carbides, grain growth results in austenite grains having a grain size in excess of 5 μm. In addition to the high m value obtained from change-in-strain-rate tests for the UHC-3Si steel (Fig. 2), high tensile elongations were obtained from constant cross-head speed tests. Even

at high strain rates, e = 100% per minute ($1.7 \times 10^{-2} s^{-1}$) at $800^{o}C$, elongations of about 500% were obtained. An additional benefit of the newly developed UHC-3Si steel is that it has a low resistance to plastic flow upon deformation in the superplastic range at low strain rates (e.g., 2000 psi at e = $10^{-4} s^{-1}$ at $800^{o}C$). This means that a UHC steel, prepared as a fine grained sheet, can be blowformed superplastically with existing, commercial, blow-forming equipment. In contrast, the superplastic UHC steels made previously were five times stronger than the UHC-3Si steel in the superplastic range (e.g., 10,000 psi at $\dot{e}$ = $10^{-4} s^{-1}$ at $650^{o}C$).[13]

1.2 Extending Superplasticity to Cast Irons

Superplasticity has been developed in compositions containing even more carbon than UHC steels, i.e., white cast irons. These white cast irons have been made fine grained by one of a number of powder metallurgy techniques. These techniques include liquid atomization[24] or rapid solidification technology (RST) processing.[26,27] Such techniques lead to powders which, upon annealing at low temperature (600-700 ^{o}C), have the desired fine micro-structure[24-29] for superplasticity. The powders are readily compacted into fully-dense compacts by warm pressing at temperatures below the A_1 or by multiple phase transformations, under pressure, through thermal cycling. Because the compaction temperature is low, ultrafine structures (1-2 μm ferrite grain size) are found in densified compacts. These white cast irons are superplastic at intermediate temperatures. A maximum tensile elongation of 1410% has been found for a 3.0%C + 1.5%Cr white cast iron.[26]

2. Novel Processing Techniques to Develop Ultrafine Microstructures in UHC Steels

Major improvements in the thermomechanical processing of UHC steels have been made. These recently developed methods[33-35] all incorporate a Divorced Eutectoid Transformation (DET) step. As a result, the total strain required to develop a fully spheroidized structure is significantly reduced. Furthermore, the need for isothermal processing, a necessary part of the former procedures, is eliminated.

The microstructure of UHC steels that have been slow cooled from a temperature in the single phase austenite field consists of a continuous layer of proeutectoid cementite, at prior austenite grain boundaries, surrounding pearlite colonies. As a result of the carbide network, the mechanical properties of steels having such microstructures are poor. In order to develop ultrafine microstructures of spheroidized cementite in fine grained ferrite, which possess excellent mechanical properties, two microstructural changes are necessary. The first change requires the formation of discontinous, fine, spherical, proeutectoid cementite particles and the second requires spheroidization of the cementite in pearlite.

In order to bring about the first microstructural change, a hot and warm working (HWW) procedure is used and this is shown schematically in the left hand side of Fig. 3 in the form of a temperature versus strain plot. In this first step, the UHC steel is initially heated to a temperature in the single phase austenite region for a sufficient time to dissolve all the carbon. The steel is then deformed during cooling to a temperature in the vicinity of the A_1 transformation temperature (727°C). Because the austenite grain size is significantly refined in this HWW procedure, the normally massive proeutec-

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toid cementite is now precipitated in relatively fine form at austenite grain boundaries as well as within the austenite grains at high dislocation density sites. The matrix of the steel consists of pearlite formed by the transformation of austenite of eutectoid composition. The microstructure of such a UHC steel after HWW is shown in Fig. 3(A). After the HWW procedure the UHC steel is austenitized at a temperature just above the A_1 temperature for a relatively short time and then allowed to cool in air. A fully spheroidized structure is obtained as shown in Fig. 3(B). The structure arises from a Divorced Eutectoid Transformation (DET). In a DET, pearlite formation does not occur upon eutectoid transformation; instead, carbide precipitation in the form of spheres takes place at undissolved carbide sites at austenite grain boundaries and within the austenite grains.

In a variation of the process described above, UHC steel in the HWW condition is heated to just above the A_1 temperature and then is deformed during cooling. In this case, the Divorced Eutectoid Transformation occurs With Associated Deformation (DETWAD). Fully spheroidized structures are also observed after DETWAD processing. Structures developed by a DETWAD process are generally finer, especially in ferrite grain size, than those developed using DET.

The microstructural changes that have been observed to take place in the DET and DETWAD experiments can be explained by using the schematic model illustrated in Fig. 4. For reference, the Fe-C phase diagram is shown adjoining the figure. In Fig. 4A, a schematic HWW microstructure is shown. The structure consists of spheroidized, proeutectoid cementite in a pearlitic matrix. A carbon concentration profile, corresponding to the location of the dashed line drawn through the microstructure, is also shown. Figures 4B and 4C illustrate the manner in which the carbides dissolve during the austenitizing

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heat treatment. The supply of carbon to form austenite comes from two sources - not only from the cementite plates in the pearlite, but also from the spheroidized proeutectoid cementite. This latter contribution from the proeutectoid cementite is not available in austenite formed in steel of eutectoid and hypoeutectoid compositions and is the principal factor that makes DET and DETWAD possible in UHC steels. In Fig. 4C, austenitizing is just complete, i.e., no ferrite remains in the microstructure. At this point, some of the carbide from the cementite plates in the original pearlite remains in the microstructure. This carbide is now classified as proeutectoid cementite. Although austenitizing is complete, the carbon is still inhomogeneously distributed in the austenite as shown in the carbon concentration profile. The structure in Fig. 4C is the optimum one for forming a DET structure upon a cooling. This is because there is a large number of nucleation sites available for carbide precipitation from austenite. These sites are the closely spaced proeutectoid cementite particles. In addition, the high carbon austenite regions may provide additional nucleation sites. Long soaking times during austenitizing lead to the microstructure shown in Fig. 4D. A uniform concentration of carbon is achieved in the austenite, and proeutectoid carbides grow by Ostwald ripening so that the interparticle spacing is increased. In this case, DET will not occur because of the reduced number of available nucleation sites and therefore the normal pearlite reaction will take place.

The economics of processing UHC steels can be considerably enhanced by utilizing DET and DETWAD procedures. One example would be to use continuous annealing line (CAL) methods recently developed by Japanese companies for processing high strength sheet steels. Thus, CAL processing of a hot and warm rolled UHC steel sheet would be an economical method of obtaining a fully spheroidized, strong and ductile steel.

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3.   Mechanical Properties of DET and DETWAD Processed UHC Steels

UHC steels processed by the novel processing routes (HWW plus DET and HWW plus DETWAD) show excellent tensile properties at room temperature. For example, in Fig. 5, stress-strain curves are shown of a 1.5%C UHC steel (1.5%Cr, 0.5%Si and 0.5%Mn, balance iron) that was processed by HWW plus DET processing and by HWW plus DETWAD processing. For comparison, the same figure also includes stress-strain curves for three commercially-available steels: a mild steel (SAE 950X), a HSLA (High Strength Low Alloy) steel (SAE 980X), and a dual phase steel (GM 980X). [The gage length of the tensile samples used to determine the stress-strain curve of the UHC steels was 0.5".] The stress-strain curves for the UHC steels demonstrate that the tensile properties of the steels are potentially better than those in the commercial steels. Specifically, the DET material shows a yield strength of 640 MPa (92 ksi) and a ultimate tensile strength (UTS) of 850 MPa (125 ksi). These strength values are higher than those for the three commercially-available steels. For example, UTS values for the mild steel, HSLA steel and dual phase steel are 450 MPa (65 ksi), 610 MPa (88 ksi) and 640 MPa (92 ksi), respectively. Furthermore, the elongation-to-failure for the DET material is about 35%. This value is also comparable to or larger than those for the three commercially-available steels. These data suggest that the 1.5%C UHC steel, processed by the HWW plus DET route, has better strength, with accompanying high ductility, than any of the three commercially-available steels shown in Fig. 5. The DETWAD processed material shows a higher strength (UTS of 1100 MPa (160 ksi)) than the DET processed material, but slightly lower ductility (about 20% elongation-to-failure).

An overview of the available tensile properties of UHC steels, achieved by the new processing routes, are presented in Fig. 6. In this graph, the

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tensile strength is plotted as a function of the tensile ductility at room temperature for four groups of steels: recovery - annealed or quenched mild steels, HSS (high strength steels) steels, dual phase steels, and UHC steels. The UHC steels shown in Fig. 6 include 1.0%C and 1.3%C plain carbon steels, a 1.25%C UHC-3Si steel, and the 1.5% UHC steels shown in Fig. 5. The properties of UHC steels, on the basis of this strength-ductility comparison, appear to be an improvement over the properties attainable in traditional mild steels, HSS steels, and dual phase steels.

4. Effect of Heat Treatment on the Room Temperature Tensile Properties of Fine-Grained UHC Steels

The room temperature tensile properties of UHC steels have been studied as a function of a variety of heat treatments.[36] The initial step in the heat treatment consists of heating the fine grained UHC steel to a temperature within the austenite plus cementite range (e.g., 770°C) for austenitizing. Upon heating, ferrite transforms to austenite at the A_1 temperature as cementite dissolves. However, a fine austenite grain size ($< 5\ \mu$m) is retained because fine particles ($0.5\ \mu$m) of undissolved proeutectoid cementite pin austenite grain boundares and inhibit grain growth. Upon quenching, quenching-and-tempering, or austempering, these fine austenite grains result in ultra-fine transformation products. For example, ultrafine martensite structures obtained in UHC steel sheet (5 mm thick) by quenching into water have previously been shown to have extremely high hardness, R_c = 65-68, as well as high compression ductility.[17] The more recent studies on quenching-and-tempering and austempering methods have led to ultrafine tempered martensite and ultrafine bainite, respectively.[37]

Figure 7 compares the stress-strain curves of austempered UHC steels with UHC steels in the as-warm-rolled condition. The stress-strain curve for mild steel is also shown for comparison. The austempered UHC steels are seen to have twice the yield strength of the as-rolled UHC steels. This is an impressive enhancement of strength by heat treatment. The ductility of the austempered 1%C - 1.5%Cr UHC steel is especially remarkable (about 20% elongation to failure) and this can be attributed to the high strain hardening rate observed in the material.

In Fig. 8 the ultimate tensile strength is plotted as a function of elongation-to-failure for fine-grained UHC steels after two different heat treatments. Also shown in Fig. 8 are tensile strength versus elongation-to-failure relations for the three groups of commercially available steels previously described (i.e., mild steels, dual phase steels and HSLA steels). It is demonstrated that heat-treated, fine-grained UHC steels can show better combinations of strength and ductility than many of these commercially-available steels. In particular, austempered fine-grained UHC steels exhibit good combinations of strength and ductility (Fig. 8).

5. Superplastic Solid State Bonding of UHC Steels and UHC Steel Laminated Composites

A key attribute of superplastic UHC steels is that they can be solid state bonded either to themselves or to other ferrous materials at temperatures below the A_1 temperature.[18] This property is significant for a number of reasons: (1) the low bonding temperature means that ferrous laminated composites can be prepared without destroying the desired fine structure in the UHC steel, (2) rapidly solidified powders containing fine structures can be pressed to a desired shape at intermediate temperatures, thus retaining

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the fine structure in the powder, (3) laminated composites of UHC steel can be prepared which permit selective heat treatment, e.g., the UHC steel can be transformed to martensite of exceptional hardness, leaving the other component unaffected,[18] and (4) welding of UHC steels by solid state bonding techniques, e.g., by tack welding, is readily achieved.

The results described above suggest some unique properties can be achieved by utilizing novel laminate design concepts. Two areas currently under investigation are impact properties and superplastic properties of ferrous laminated composites based on UHC steel. An example from each area is given in the following.

High impact strengths have been obtained in laminated composites, the components of which exhibit lower impact strengths than the laminate. An example of such impact behavior of a UHC steel mild-steel composite is illustrated in Fig. 9. The notch impact strength of the laminated composite is shown to be much higher than those observed either for the mild steel or for the UHC steel. Furthermore, the impact transition temperature for the laminated composite is seen to be at a very low temperature, about $-140^{\circ}C$, much below those for either of the components that make up the composite. The high impact resistance of the composite is attributed to notch blunting of the crack by delamination at the layer interfaces.[23] Such delamination is readily seen in the left hand sample illustrated in Fig. 9. The high impact resistance of the laminate composite thus is due to the presence of a good (but not perfect) bond between laminates. If the bond strength is improved, e.g., by a thermal cycle heat treatment, the impact strength is degraded.[23]

A laminated composite consisting of a superplastic and non-superplastic component can be made to exhibit superplastic behavior. A UHC steel-mild

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steel laminated composite has been shown to exhibit strain rate sensitivity exponents of over 0.30 and elongations to fracture of over 400%.[19-21] The strain rate-stress results show good agreement with constitutive equations for creep which have been developed based on an isostrain creep deformation model. The equations lead to quantitative predictions of material requirements for achieving ideal superplasticity ($m = 0.5$) in laminated composites based on UHC steel. A laminated composite consisting of a ferritic, stainless-steel-clad, ultrahigh carbon steel is predicted to exhibit ideal superplasticity at 800 $^{\circ}$C. Current studies by Daehn, Kum and Sherby[38], in fact demonstrate that this condition is achieved experimentally ($m = 0.5$ and elongation to failure over 800%). This combination of components leads to the unexpected result that coarse-grained stainless steels can be made superplastic.

Conclusions

It is shown that ultrahigh carbon steels can be readily processed to possess highly desirable mechanical properties: superplasticity at intermediate temperatures, and high strength and high ductility at low temperatures. Thermal-mechanical treatment procedures for achieving fine spheroidized structures in these steels have been developed which are amenable to mass production techniques. It is timely to consider the initiation of development studies on UHC steels and to evaluate their properties on prototype structural components.

Acknowledgments

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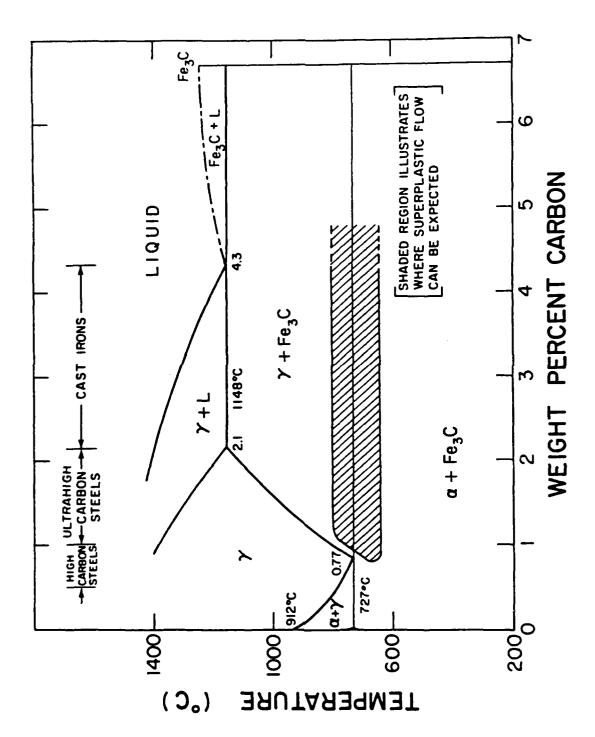
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Figure 1. The iron-carbon (cementite) phase diagram is shown. The shaded area illustrates the temperature and composition range over which ultrahigh carbon steels and white cast irons have been made superplastic.
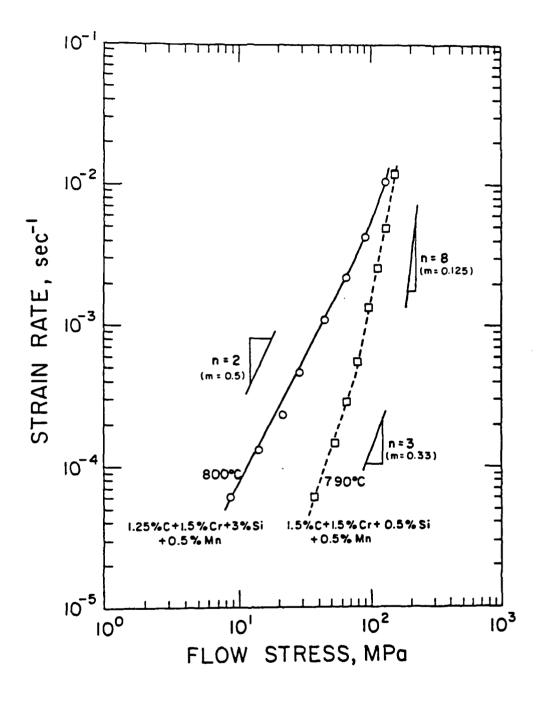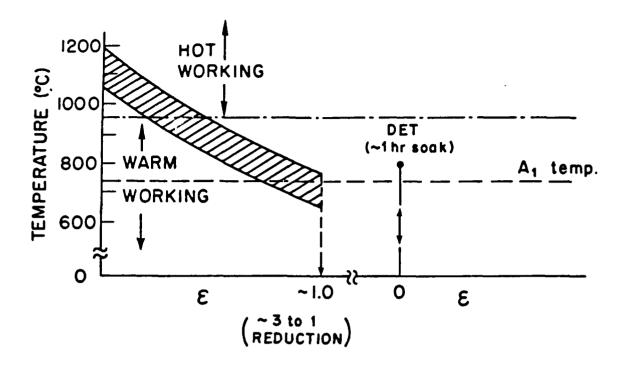
Figure 2. Logarithm of the strain rate as a function of the logarithm of the flow stress for the UHC-3Si steel (at 800°C) and for the low-Si UHC steel (at 790°C).
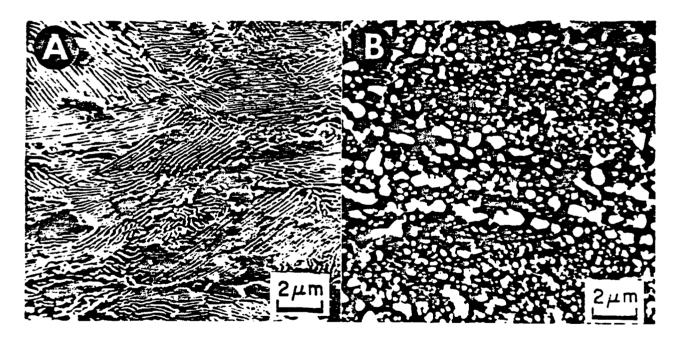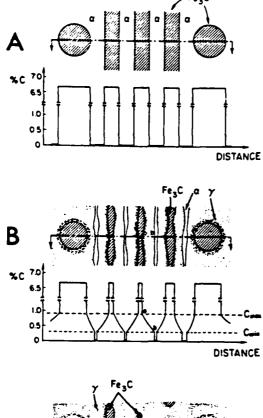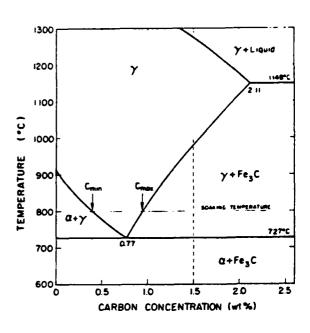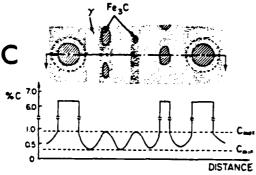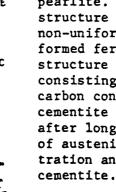
Figure 3. Schematic illustration of a hot and warm working (HWW) process plus a divorced eutectoid transformation (DET) process. Scanning electron micrographs show microstructures (A) after Stage 1 (HWW) processing, and (B) after Stage 2 (DET) processing of a UHC steel containing 1.5%C, 1.5%Cr, 0.5%Mn, 0.5%Si and balance iron.
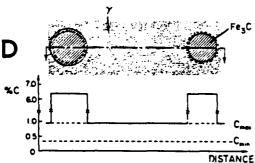
Figure 4.
Schematic illustrations of the stages during austenitizing of a UHC steel after Hot and Warm Working (HWW). A carbon concentration profile for each stage is also presented. A, initial HWW structure consisting of spheroidized proeutectoid cementite and pearlite. B, incompletely austenitized structure consisting of austenite with a non-uniform carbon concentration, untransformed ferrite and cementite. C, a microstructure just after complete austenitizing, consisting of austenite with non-uniform carbon concentration and (proeutectoid) cementite particles. D, microstructure after long-time austenitizing consisting of austenite with a uniform carbon concentration and large particle proeutectoid cementite.
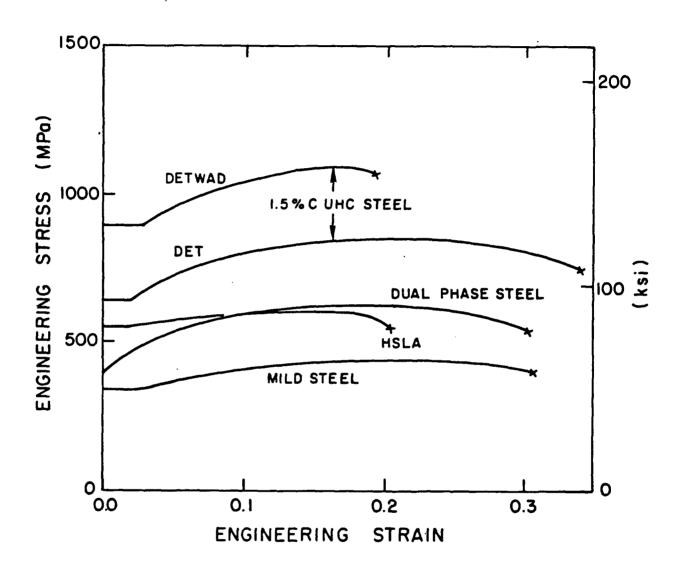
Figure 5. Engineering stress – engineering strain curves at room
temperature comparing a UHC steel after DET and DETWAD
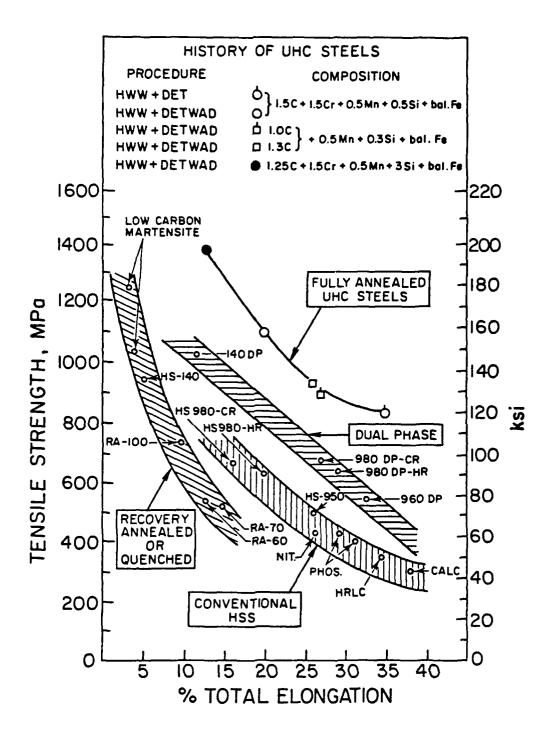processing with a mild steel, an HSLA steel and a dual
phase steel.

Figure 6. The tensile strength and percent elongation to fracture of UHC steels at room temperature is compared with mild steels, high strength steels and dual phase steels.
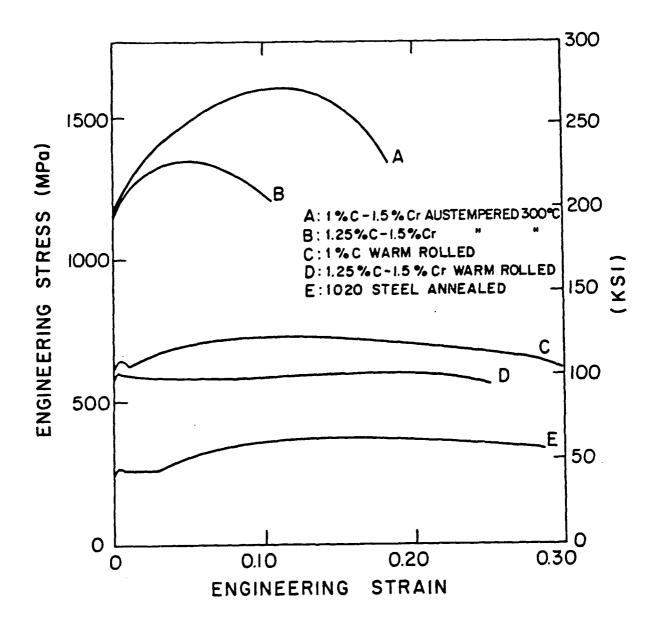
Figure 7. Stress – strain curves for austempered and as–warm-rolled
fine-grained UHC steels. A stress – strain curve for 1020
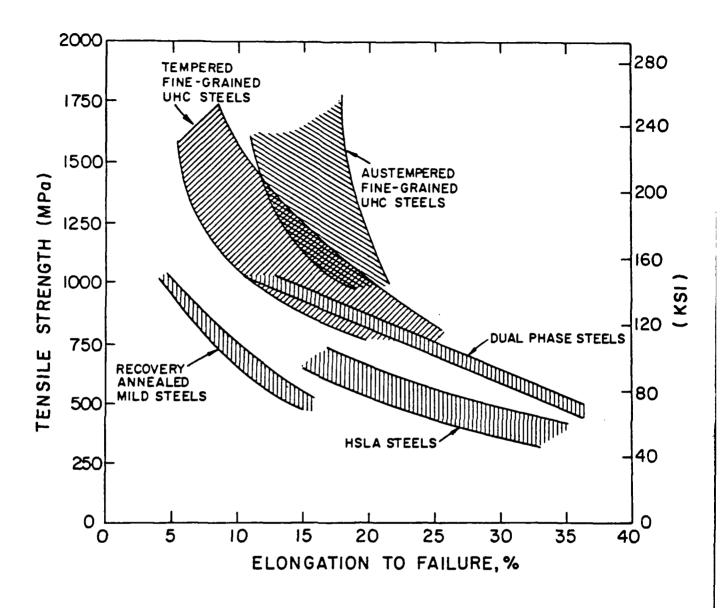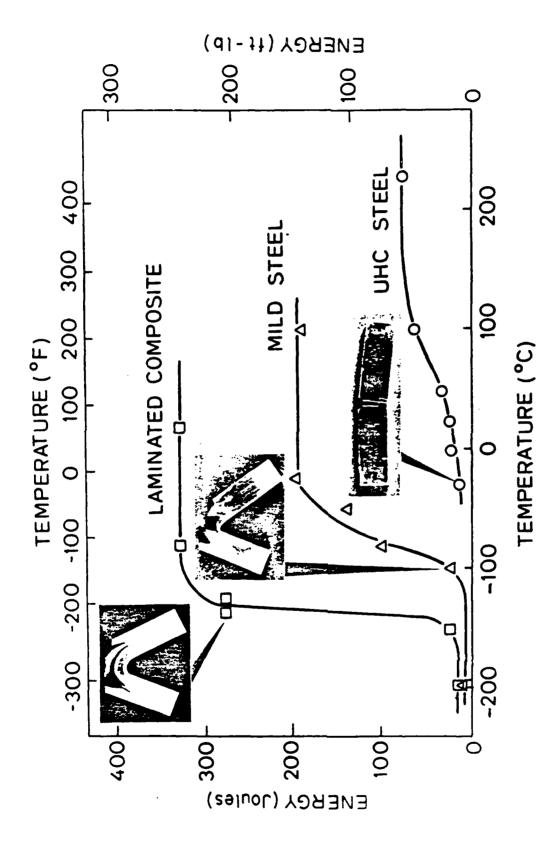steel is included for comparison.

Figure 8. Tensile strength and elongation to failure of fine-grained UHC steels compared to mild steel, HLSA and dual phase steels.

Figure 9. Influence of temperature on the impact properties of a laminated composite of UHC steel and mild steel compared with the component materials making up the composite. The mode of fracture is shown with the insets given in the figure.

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